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Toruń, 10.08.2022.

**Review Report on the PhD thesis submitted to Thomas Bata
University in Zlin, Czech Republic by Hau Trung Nguyen
Title: Kombucha leather: Preparation and Characterization
Suprvisor: Assoc. Prof. Nabanita Saha, Ph.D.
Consultant: Prof. Ing. Petr Saha**

The presented review report is organized in the following sections: project background, general description of the thesis, specific comments followed by a final evaluation statement.

Project background

Genuine leather is a strong and durable material which is usually obtained from animal skin. To get good quality leather, a chemical treatment is required, called the tanning process. During the tanning process, the rawhide is converted into a stable, pliable, residue-free material suitable for a wide range of finish applications. To obtain good quality leather, different tanning agents can be used and depending on the procedure, several types of leather such as vegetable-tanned leather, chrome leather, aldehyde leather, can be produced. However, the genuine leather processing caused a multitude of wastes from animal meat, feathers, debris, exhaust gases, and volatile solvents to numerous wastewater containing harmful chemicals such as chromium, tannins compounds, oils, resins, biocides and detergents. In several countries, wastewater from tanneries is a big problem and has been subject to general industrial discharge laws or an obligatory standard. The next problem is agroforestry by-products or bio-waste, which need to be reduced. The reuse of agroforestry by-products or bio-waste is one of the possible approaches that scientists and producers priority selected to perfect alternative leather materials, notably vegan leathers, including fruit leathers, waxed cotton leathers, button leather, wood leathers, ocean plastic leathers, mushroom-derived leathers, bacterial cellulose-derived leathers or kombucha leathers.

Kombucha leather is a bacterial cellulose-derived leather, a type of vegan leather that resembles leather but is not made from animal skin. This kind of leather is of high social expectation, with high novelty, is environmentally friendly, using less energy consumption. It gives less harmful emissions and requires very little space, water, or chemicals to produce. As currently, sustainable and environmentally friendly production are critical points for the future of the leather industry, the PhD thesis prepared by Hau Trung Nguyen regarding the preparation of leather-like biocomposites based on kombucha-derived bacterial cellulose (KBC) is timely and may offer new ideas and materials for leather production. The leather industry is constantly shifting to vegan leather or eco-leather due to the continuous increase in awareness of consumers and manufacturers. It is connected with regulatory agencies regarding ethical consumption, cleaner production, sustainable development, and animal rights. Moreover, it can be a well-response to this urgent requirement of the leather industry and may influence the future of leather markets.

The PhD thesis of Hau Trung Nguyen is focused on the development of kombucha leather that can have a possible application in fashion design, footwear, bags, and interior covering products. It has been achieved by a combination between polyurethane elastomer, polylactic acid and KBC harvested from bio-wastes fermentation.

General description of the thesis

The submitted thesis presents investigations on the preparation and characterization of kombucha leather by a combination between polyurethane elastomer, polylactic acid and KBC harvested from the fermentation of bio-wastes. The incompatibility between the hydrophilic bacterial cellulose (BC) and the hydrophobic polymer matrix, which causes easily broken structures and highly water absorbents, was overcome by pretreatment KBC/BC with silane compounds.

The dissertation comprises 116 pages (plus pages containing published articles and Annexes with the scientific outcome of the Author). It does not follow the classic structure of a PhD thesis in science. It begins with abstracts (in English and Czech) and an introduction to the subject, which is called Background. In this part, the literature review on genuine leather production, bacterial cellulose (BC) and kombucha-derived bacterial cellulose (KBC) has been done. The literature for this part is carefully referenced (210 references), and a comprehensive bibliography contains all the relevant papers for the discussed field. It is worth mentioning that most of the references are from the last decade, showing the topical issues. This part is well written and provides a perfect background for understanding the rest of the thesis.

The main part of the thesis – Part II Experimentation – consists of four chapters (Chapters 2.1-2.4), each addressing a specific method used in the research. Chapter 3 is dedicated to the motivation for the doctoral thesis, whereas chapter 4 shows the aim of the doctoral thesis and its summary. The results, which are the basic part of the doctoral thesis have already been published, and the scientific papers have been included in the PhD thesis. The results have also been presented at scientific conferences and published in conference proceedings. The aim of the thesis and its objectives are clearly stated.

The work concerns four objectives.

1. Effective evaluation of KBC production using media containing sour whey waste, waste apple juice, and brewed spent grains.
2. Optimization of KBC production and trial of the result in large containers.
3. Influence of silane compounds on the hydrophobicity of KBC and determine usable polymers for kombucha leather preparation.
4. Optimization of kombucha leather preparation.

The obtained kombucha leather products were compared with commercial leathers in structural, morphological, thermal resistance, surface wettability, and mechanical strength to evaluate their applied potential. The details of this doctoral study have been published in four peer-reviewed journals, which are indexed on the Web of Science, and one AIP conference proceedings, as well as one utility model of the Czech Republic. As already published, all the papers met the requirements of the journals and underwent assessment by the reviewers of the journals, so there is no need to assess the published papers again.

The following publications (4 assessed already by journal reviewers) form the results part of the doctoral thesis.

Publication 1:

Kombucha-derived bacterial cellulose from diverse wastes: a prudent leather alternative. Hau Trung Nguyen. Nabanita Saha. Fahanwi Asabuwa Ngwabebhoh. Oyunchimeg Zandraa. Tomas Saha. Petr Saha. *Cellulose*, (2021), 28, 14, 9335-9353. Q1, IF 5.044.

Publication 2:

Development of novel biocomposites based on the clean production of microbial cellulose from dairy waste (sour whey). Hau Trung Nguyen, Fahanwi Asabuwa Ngwabebhoh, Nabanita Saha, Oyunchimeg Zandraa, Tomas Saha, Petr Saha. *Journal of Applied Polymer Science*, (2021), e51433. Q2, IF 3.125.

Publication 3:

Preparation and characterization of nonwoven fibrous biocomposites for footwear components. Fahanwi Asabuwa Ngwabebhoh, Nabanita Saha, Hau Trung Nguyen, Urška Vrabič Brodnjak, Tomas Saha, Anežka Lengalova, Petr Saha. *Polymers*, (2020), 12, 3061. Q1, IF 4.329.

Publication 4:

Silane modified Kombucha cellulose-based biocomposite leather-like mats: Preparation, optimization, and characterization. Hau Trung Nguyen. Nabanita Saha. Fahanwi Asabuwa Ngwabebhoh. Oyunchimeg Zandraa. Tomas Saha. Petr Saha. Manuscript is submitted to *Sustainable Materials and Technologies* on 01/04/2022 (status: under review in June 2022).

Publication 5:

Environmentally Friendly and Animal Free Leather: Fabrication and Characterization. Nabanita Saha, Fahanwi Asabuwa Ngwabebhoh, Hau Trung Nguyen, Petr Saha. *International Polymer Processing Society Europe - Africa 2019*, Regional Conference, November 18 – 21, 2019, Pretoria, South Africa. AIP Conference Proceedings 2289, 020049 (2020).

Publication 6:

Leather material with improved ecological parameters. Utility model. CZ 33149 U1. Fahanwi Asabuwa Ngwabebhoh, Nabanita Saha, Hau Trung Nguyen, Tomas Saha, Petr Saha. *Industrial Property Office, Czech Republic* (2019).

It is worth mentioning that the journals mentioned above are of high scientific quality, with high IF, ranking in Q1 and Q2.

Each publication solved a specific scientific problem. All of them exhibit the same structure and are divided into sections, which are typical for scientific papers: introduction, experimental, discussion, conclusions and references. The advantage of such a structure is that each publication can be read and analyzed separately, but finally, the mentioned publications form an integral and well-organized PhD thesis. It is also a small disadvantage because some portions of information found in the introductions, experimental sections and discussions are repeated. In the final section, the general conclusions of the work are summarized. The thesis is prepared with good editing standards. All the figures are carefully prepared and well presented. The language is comprehensive and coherent, while errors and inaccuracies are relatively rare.

Specific comments

The title of the thesis, “Kombucha leather: Preparation and Characterization”, seems to be too general and does not show all the aspects of the doctoral dissertation. I understand that the kombucha leather was the main material prepared within the thesis; however, I have found much more important information in the thesis, which are worth to be mentioned in the title. The experimental part is clearly written, but sometimes more information is required to understand the scientific work.

Concerning the experimental part of the thesis, I would like the candidate to answer the following minor points during the defence:

1. The mechanical properties of materials may be dependent on relative humidity during the measurement. What was the relative humidity, and how could it influence the received results?
2. Biodegradation studies were performed in a humidity chamber with temperatures maintained at 30 °C and 50% surrounding humidity. What was the rationale for such conditions?

In general, the presented results seem to be reliable and well documented. I would like to emphasize that the studies concern a broad area of research, and their main value consists in the detailed description of the obtained results and potential future applications.

The summary and general conclusions.

The main goals of this thesis were quite ambitious, but the results showed, that they have been achieved to a large extent, as demonstrated in published papers. The Author correctly highlights the contribution of the thesis in the current science regarding leather production and also notices its limitations, showing clearly that the PhD student has a comprehensive understanding of the context of his work. As the final outcome of the doctoral thesis kombucha leather was prepared and successfully characterized. Optimized kombucha leather material containing PU, PLA, and KBC pretreated with vinyltriethoxysilane was animal-free, with a moderated rate of biodegradation and good elasticity and a hydrophobic surface. The comparison between optimized kombucha leather and two commercial leather types showed that the basic requirements of this new leather substitute material had been met.

Final evaluation statement

This thesis represents a great deal of work. The results are well presented, and their interpretation is at a high scientific level. I do appreciate the candidate's expertise in the field of materials characterization and BC preparation. The research described in the thesis is of the international standard. This thesis is ready to be defended orally and certainly meets the requirements for the degree of PhD in material science and chemistry according to the rules of Thomas Bata University in Zlin, Czech Republic. As an External Reviewer of the above-mentioned doctoral thesis, I recommend it for the defence in its current form.

